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PROPOSED TECHNIQUE ON SPENT FUEL DISPOSAL

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ABSTRACT

The spent fuel elements from nuclear power stations are reprocessed after a period of storage, and then the residual wastes are converted into solid blocks through vitrification technology. Low-level radioactivity of spent fuel demanded waste management for deep sea disposal and also the inceneration technology. Intermediate level radioactive wastes from spent fuel require containment in an engineered storage system before finally disposing of them to sea.

In this paper, we are mainly concerned with high level waste management practices, vitrification technology, transportation and disposal of wastes. Our proposed technique on spent fuel disposal require deep drilling in larger areas of granite and similar hard rocks covered and surrounded by clays and salt formations. Naturally, the folded structure of hard rocks surrounded by areas of salt formations would lead to a permanent storage for high level radioactive wastes.

1. INTRODUCTION

There are two generic disposal methods: (i) dilution and controlled dispersion to the environment; and (ii) concentration and burial or storage. High-level liquid wastes and solid waste matter are invariably stored or buried. The terms low-, intermediate - and high-refer to the concentration of radioactivity. We endorse, in particular, the expansion of the geological and oceanographic research programmes for high-level waste disposal.

The major source of waste from nuclear industry is the spent fuel elements from nuclear power stations. These contain unburnt uranium, plutonium and waste products arising from nuclear interactions within the fuel in the reactor. After a period of storage, the spent fuel is reprocessed to seperate out these various components, permitting the unburnt uranium and plutonium to be extracted and recycled for further use. The residual wastes from reprocessing are highly radioactive and form the high-level nuclear wastes. Storage is not a substitute for disposal, although it is a necessary part of waste management. The transition from storage (containment in an engineered storage system) to disposal is unlikely to take place until at least 50 years after the wastes have been vitrified, assuming vitrification will take place at least five years after the removal of fuel from the reactor. We are presently concerned with optimisation of waste storage and disposal. Further research is needed to ascertain the disposal route which is best from the point of view of safety, economy and public acceptibility. The duration of the subsequent period of storage in vitrified form depends on a number of factors, the most important of which is the eventual method of disposal to be adopted; a shorter period may be required if the waste is disposed of on the deep ocean bed then if it is buried below the sea bed or the land surface.

The containment in an engineered storage system, either above ground or sub-surface - for which technology already exists might be the best way to deal with solidified high-level wastes for at least 50 years and possibly much longer. In examining the waste management implications of the nuclear power programme, we have based our calculations on the figure of 25 GW which is the most recent estimate of likely installed nuclear generating capacity in the year 2000. In the case of high-level radioactive waste remaining from the reprocessing of spent fuel, the envisaged increase in capacity is unlikely to present any signigicant new waste management task. The total high-level waste arising up to the year 2000 from a 25 GW programme would be less than 600 m^3 in volume. The type of reactor is unlikely to alter the activity, or significantly affect the volume, of high-level vitrified waste. Geological disposal is only one of a number of possible methods of dealing with high-level wastes.

J.B. Lewis (1) summarised with arguments the unsuitability of dumping the high-level radioactive wastes in the ocean beds, particularly, due to problems arising from monitoring the effects caused by deep sea disposal. The principal natural source of internal radiation to deep sea fish is Po 210 and a study of this in fish provides a useful indication of how other radionuclides will behave. However, a more eloborate research programme is needed which includes three-dimensional waste dispersion model, two-dimensional meridional model and process models. These models will provide informations about accurate estimates of concentration fields, natural trace distributions and physical transport processes in specefic locations.

2. REASONS FOR NUCLEAR FUEL REPROCESSING

Fission products contained in irradiated fuel elements constitute the dominant radioactive waste disposal problem in nuclear industry. High-level wastes include the isotopes of neptunium (Np), americium (Am) and curium (Cm), alongwith small amounts of uranium and plutonium, that would not be removed in reprocessing owing to ineffeciencies in chemical seperation. When reprocessing has been carried out, the high-level radioactive waste is initially in liquid form. During fuel reprocessing operations, the fission products are freed from the normal confinement of fuel element cladding. Before, being transported to a reprocessing facility, irradiated fuel elements are stored in a water-filled basin for about 3 months of take advantage of the initially high rate of decay of fission products activity. The cost of constructing tank forms for high-level radioactive waste storage depends on site conditions and on the chemical nature of the waste solutions.

Hundreds of different fission products are formed in a nuclear reactor, of which strontium 90 and cesium 137, two characteristic fission products, constitute about 5% of the total, particularly, in LWR (light water reactors). The fissionable plutonium 239 is consumed by fission reactions, near the end of the effective life of the fuel, as fast as it is being created. The effective life of the fuel is determined from the design characteristics of fuel composition and reactors. However, the fissionable plutonium is bred in some fast reactors. In this particular case, we produce plutonium about twice than what we consume. From the time spent fuel is removed from the reactor, it consists of fission products which includes unstable nuclides such as xenon 135 and cesium 135 which decays fast to various other elements, stable nuclides such as Sm 149 etc, unburnt isotopes of uranium and plutonium, cladding and engineering materials.

Before reprocessing the spent nuclear fuel, sometimes post-irradiation processing is carried out for the enrichment of unranium and plutonium. This is accomplished by adding uranium and plutonium of which the enrichment is required, in the fission products and then a particular chemical engineering process known as "Purex Process" is followed.However, we should have a plant design philosphy of "Purex Process" to achieve our cause for a specefic purpose.

Now, we briefly describe the following reasons and its objectives for spent fuel reprocessing.

(1) - Physical and mechanical changes in the properties of the fuel.

(2) - Reactivity of fuel decreases with time due to growth of fission product poisons.

(3) - Plutonium is a valuable byproduct of fission. It should be recovered for the use in fast breeder reactors.

(4) - To seperate out low-, intermediate-, and high-level radioactive wastes and to propose methods for their disposal.

Objectives:

(1) - To recover Pu in pure form with >99.9% effeciency.

(2) - To reduce uranium content of plutonium to <1%.

(3) - To reduce fission product activity in Pu by a factor of $\sim 10^6$

(4) - To recover depleted uranium in pure form from fission products.

(5) - To arrange fission products for permanent storage according

to activity.

(6) - To control the emission of fission products to atmosphere and effluents and make it safer for biosphere.

3. HIGH LEVEL RADIOACTIVE WASTE MANAGEMENT

High level radioactive waste for considerable length of time was already known to be practicable, and in a vitrified form would require far less supervision than in liquid form. It is easier, safer and cheaper to contain and store solids than liquids and it is logical to convert the waste into solid blocks. While tank storage of high-level liquid waste is manageable on a short term basis, it is not an attractive permanent solution to the disposal problem in view of the long radioactive half life of some fission nuclides. Disposal of high level radioactive wastes by dilution and dispersion is not an attractive solution in view of the large waste volumes involved and the stringent tolerances on strontium 90 concentration in the environment. The most troublesome solid wastes come, not from the reactor, but from the fuel reprocessing plants. Long before the year 2000, it will have become routine for the high-level radioactive waste produced in reprocessing fuel from these plants to be converted to solids and buried or stored where it cannot reach the biosphere.

For spent fuel and vitrified waste management, interim storage is an essential part of the cycle. The passive air-cooled vault store developed by GEC Energy Systems Limited provides an optimum engineered solution, available now for power station or reprocessing site application. Temporary storage is particularly important with respect to an isotope such as iodine 131 ($t_{\frac{1}{2}}$=8 days). For fuel management scheme, cesium discharge reduction is also an important part of the spent fuel cycle. The best method of reducing the cesium discharges in the interim would be to immerse boxes of spent ion exchange material in storage pond water at the site. The fuel, which is not normally canisterised, can be inspected or retrieved at any time. After a few years cooling in the pond, oxide fuel can be maintained safely in an air storage environment. So far the solutions they have come up will have been merely temporary, and almost twenty thousand tons of deadly waste are now scattered over the globe in steel rods housed in storage tanks and boric acid pools that are fast becoming overcrowded. By 2000, the tonnage of spent fuel may have increased five-hold, raising the storage problem to crisis proportions. Vitrification of high-level radioactive waste soon after reprocessing has been carried out, would the temperature of the blocks to rise if they were left to themselves. The temperature increase would depend on their size, shape, waste content and surroundings, but it is undesirable to allow too great a rise in temperature of the blocks because this could affect their ability to contain the radioactive wastes. In practice, it is quite easy to design storage, on or near the surface, to provide shielding and containment of the radioactivity, and to provide cooling by water or air to take away the heat generated. The amount of heat generated by the blocks decreases with time because of the reduction in radioactivity. Our recent estimates show that we should store the high-level radioactive wastes for at least 50-100 years, and may be considerably more, before the heat generation falls to a level at which the economic and simple method of disposal is possible. Professor Ringwood at Canberra has suggested that to overcome the risk of glass blocks getting hot there would be merit in using an artificial rock called Synrock (ceramic) instead of glass to contain the radioactive wastes.

Julian Weiss (2) has mentioned that there are promising methods available for the disposal of hazardous wastes (that can remain active for anything between one thousand years and millions of years), which include changing decaying matter to glass or blending it with concrete, or transforming the substance to ceramics. In a multiple barrier technique, host rock (either salt, basalt or granite) is selected. Crushed rock is used to coat metal and steel barriers that form concentric circles, while waste products are kept at the center. Monazite (phosphate molecules) could be used to contain harmful radioactive wastes for hundreds of millions of years, and found completely water resistant. One advantage in this case is that the conversion to monazite could take place at atomic energy plants, thereby avoiding transport of wastes. Moreover, a variety of borosilicate glasses can be constructed from nuclear waste. The option of vitrifying nuclear waste-drying it to powder, mixing it with glass making materials called frit, then melting and cooling into solid form-has been taken up by several countries.

A variety of different storage techniques and disposal medium exists, suited to different types of waste spent fuel or reprocessing - and the geology of different countries. The spent nuclear fuel is encapsulated in titanium, stainless steel and copper canisters, respectively. The best method of encapsulating spent nuclear fuel in copper is based on the principle of hot isostatic pressing of waste. How-ever, most of the world's nuclear byproducts are presently stored in stainless steel canisters. Two basis methods were studied by Professor B.L.Cohen (3) for storage and sidposal of radioactive wastes. These include (a) immobilization of the wastes by converting them to more easily stored solid form, or by selective fixation of the radioactivity in inert, non-leachable, and hence buriable solid material, and (b) piping the wastes into liquid form into geological formation where there is little possibility of contamination of ground water sources. Research on the former method is being conducted at a number of laboratories and involves studies of various calcination and fixation schemes. An example of one of the scheme is as follows: the waste solution is partly calcined and filtered to remove solid fission product oxides, which are compacted and packaged for storage. The residual waste solution is passed over a bed of montmorillonite clay which is effective in adsorbing fission product ions. The clay is then heated to a high temperature, becoming a refractory and essentially non-leachable material which can be buried.

The comparitively small quantities of radioactive materials involved make it practical to use highly sophisticated waste management procedures, whose cost must be viewed in relation to the price of the electricity generated. For example, a medium size, 1000 MW reactor produces thirty tons of waste each year. Ninety six percent of that waste could actually be reprocessed, thereby drastically reducing the amount of useless spent fuel for disposal, but plans for the constructions of commercial reprocessing plants have virtually come to a stand still.

As mentioned earlier that vitrification of high-level radioactive wastes demanded "cooling" in the storage before buried to any proper geological site would, however, require delayed burial, as shown in Fig (1), otherwise wastes incorporated into borosilicate glass (Similar to Pyrex) would become devitrified (crystallized or brittle) at temperatures higher than about 700C°.

4. GEOLOGICAL DISPOSAL OF HIGH-LEVEL RADIOACTIVE WASTES: METHODS AND PROPOSED MODELS:

Various proposals (4) for disposal have been made, including disposal in rocks thousands of feet underground where they would be virtually inaccessible. The best way forward for geological disposal will be to continue to cool the blocks near the surface until the heat they produce has fallen to a level at which disposal can be recommended. When we are satisfied that the isollation of such solidified wastes can be deemed permanent we call it disposal. Within the past few years, tests demonstrated the ability of vitrified glass to withstand not only heat from atomic particles but from placement deep within the earth. Large clay sediments upto 170m thick would immerse the wastes even if titanium canisters dissolved. As much as one fourth of the sea-bed is geologically stable, and if nuclear waste is mixed to form borosilicate, this type of disposal could be inexpensive and practical; but we have to consider the review article of J.B.Lewis (1) according to which dumping of high level radioactive wastes is unsuitable in deep ocean beds. The requirement that radioactive wastes be isolated for hundreds of years seems alarming to some because few things in our environment last that long. Deep underground, however, the time constants for change are in the range of 10^7-10^8 years. Thus, on any long time scale, nuclear power must be viewed as a method for cleansing the earth of radioactivity. Random burial offers less security than careful choice of a burial site based on geological information.

Professor B.L. Cohen (3) suggested few methods for additional protection aganist release during few hundred years critical, which are as follows:

(1) - The material will be buried in a geological formation that has been free of ground water for tens of millions of years and in which geologists are quite certain there will be no water for some time in future.

(2) - If water should get into the formation, the rock constituting it would have to be leached or dissolved away before water could reach the waste. Even if the rock were salt, dissolution would typically require thousands of years.

(3) - Once the water reached the waste glass, the latter would be leached at a rate of only about 1% per century.

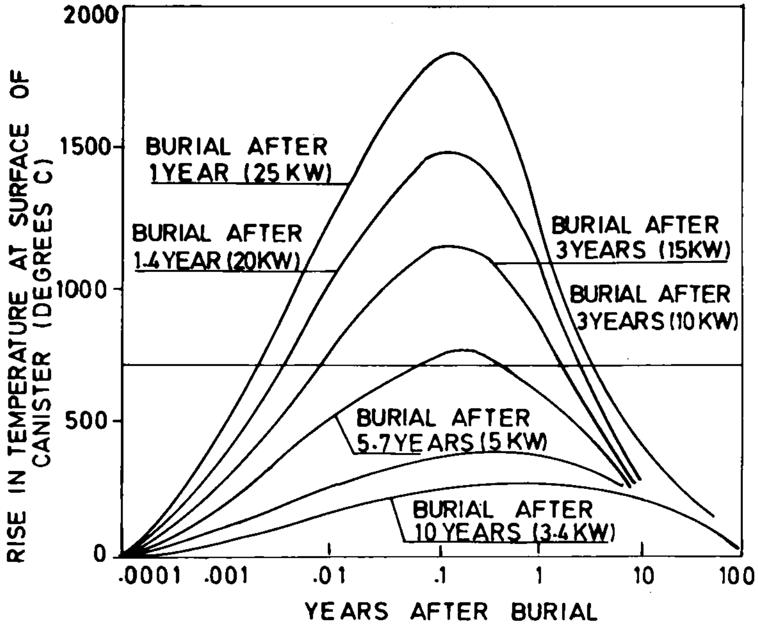

Fig. 1.

(4) - Ground water flows through aquifers rather slowly, requiring typically 1,000 years to reach surface waters from a depth of 600 m.

(5) - Most of the radioactive materials would be held up by ion exchange processes, travelling 100 to 10,000 times slower than later.

The geological formation which have been considered for storage of high-level wastes in liquid form include salt beds, deep underground basins, and excavations in selected shale formations. One of the criteria for the choice of a repository site is that there be a lack of valuable minerals and the prospect of discovering them. While discussing the various methods for the disposal of radioactive wastes, Professor B.L. Cohen (5) examined that if blocks were placed in salt under pressure, the heat would make the salt around it. This will cause the migration of the water pocket in the direction of the higher temperature, which is of course the direction of the buried waste canister. Small amount of water would continue to migrate after 25 years, carrying corrosive substances such as hydrochloric acid arising from chemical reactions induced in the salt by the radiation from the canister. While for a hard rock like granite, the heat could crack the rock and hence the migration rate of disposed high-level radioactive wastes through cracks upto the surface of the earth would become appreciably larger thus causing human intrusion.

Therefore, we suggest recommendations in future research programme for the disposal of high-level radioactive wastes, which are as follows:

(1) - Type of the rock which would be most suitable for storing the solidified wastes. Even, the placement of an artificial rock (ceramic) basin deep underground the earth's surface could be considered. This artificial rock (chemically formed ceramic material) should be compatible enough to hold high temperatures caused by buried canisters. Even if ground water comes in contact with artificial rock, this compound is tough enough to resist leakage.

(2) - An appropriate rock drilling programme to suggest various designs of the burial.

(3) - Studies of various types of deep rock formations such as on craton.

(4) - Studies on different types of rocks which have different responses to heat.

The advantage of going to depths of hundred of meters is that inactive parts of the earth's crust have geologic structures that would remain stable for millions of years. The total amount of natural radioactivity in the ground down to the proposed nuclear waste burial depth should first be determined in terms of the radium content and an allowance should be made that it should be greater than the radioactivity in the wastes under safegaurd conditions. Of course, the radioactivity of wastes is more concentrated, but in principle that does not make any difference to the biological effects of radiation. The reason behind this argument can be explored from the article of B.L. Cohen (3). Based on these arguments, we should survey and explore the amount of radium in the top 1000 meters of the earth's crust of geological stable areas of Pakistan and also the amount of radium in people.

We have discovered two design models for the geological disposal of high-level radioactive wastes, deep underground the earth's surface by considering recent theories on geological structures, which however makes it reasonable from the point of view of carrying further research. The design models if not practically suitable can, however lead to further investigations, suggestions and improvements. In the first design model, as shown in Fig (2), we have made use of the recumbent folding (dip of 0-10°) which can carry canisters of high-level radioactive wastes. The recumbent foldings are supported from the concrete walls and also from the shielding material which is made of an artificial rock (ceramic). The shielding can prevent leakage and can hold high enough temperatures caused by waste canisters. The recombent foldings as shown in Fig (2) is surrounded by salt or basalt; if water vapours developed will move towards the inclined part of the folding where we have the maximum heat due to wastes. If any chemical reaction takes place due to radiation of the wastes in salt, that corrosive substance will move towards the inclined part, and cannot be leached from the chemically formed artificial rock. This process will take place in successive steps as the heat content can be suitably transferred to the environment without causing any crack in the rock. Since folds are very common geological structures, and can be easily found in the top

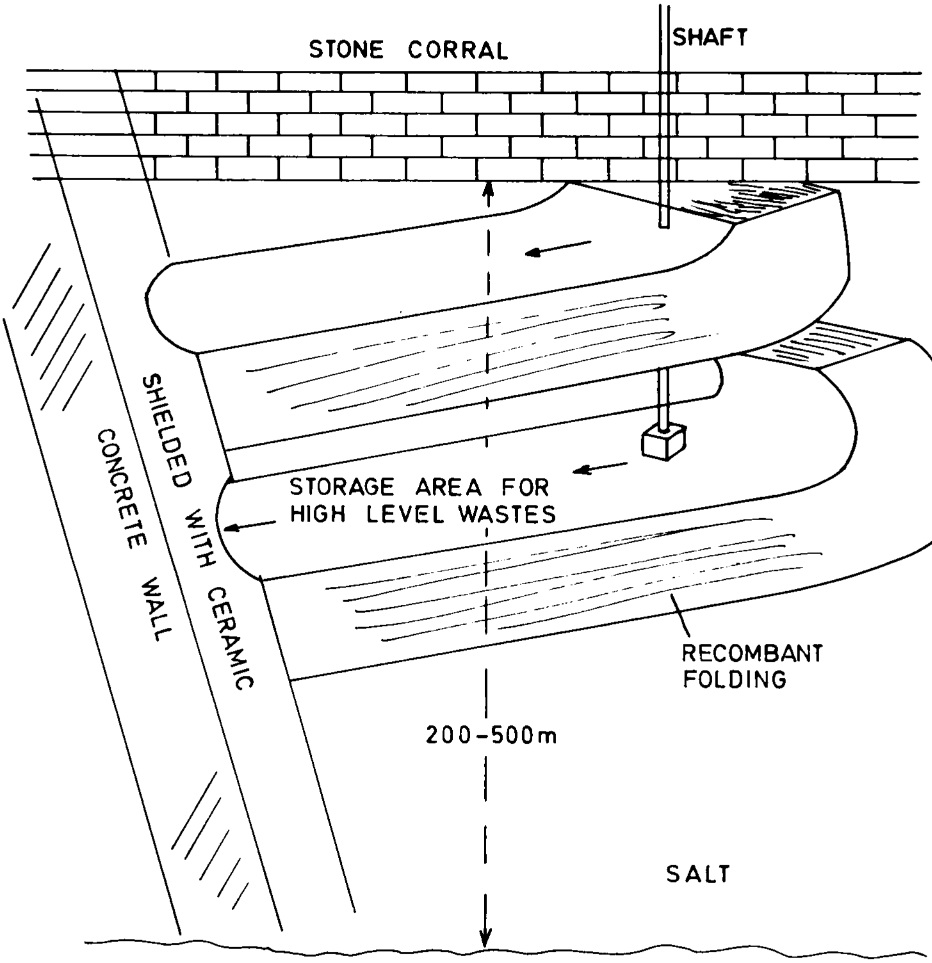

Fig. 2.

200-500 meters of the earth's crust where drilling as well as the placement of concrete and artificial rocks could be made accessible. In the second design model, as shown in Fig (3), we have considered the rock basin in the craton (which is usually 35-45 Km thick) deep underground, 400-1000 m from top of the earth's surface. The rock basin in the craton is surrounded by an artificial rock to avoid leakage of leachable materials. This method is relatively expensive than the former one and cannot be made economically viable. However, this method has the advantage of containing large amounts of waste canisters.

Since, we do not have sufficient data about the geological disposal of radioactive wastes; otherwise it is possible to simulate computer model experiments on recombent fold entirely

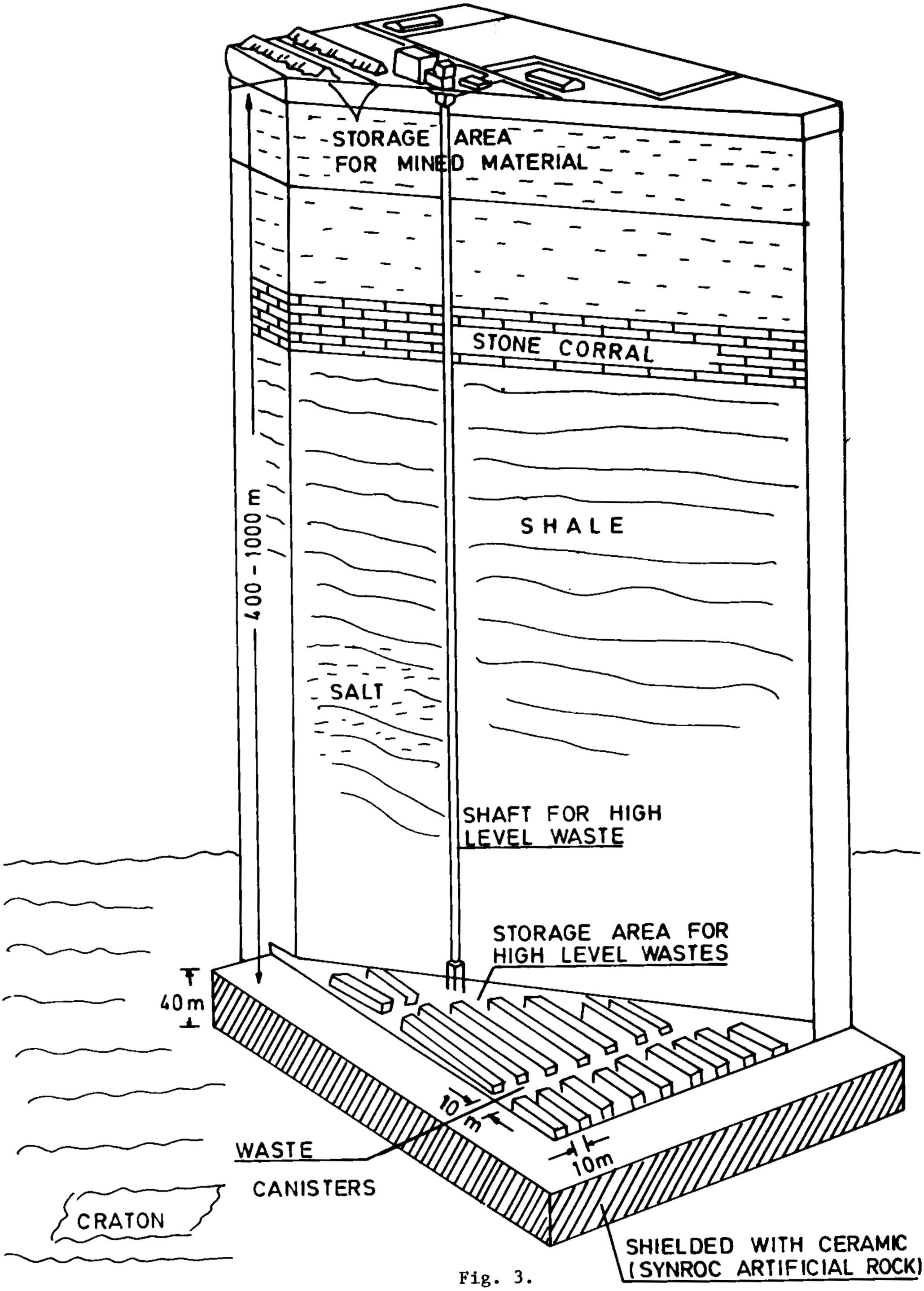
STORAGE AREA
FOR MINED MATERIAL
STONE CORRAL
400 - 1000 m
SHALE
SALT
SHAFT FOR HIGH
LEVEL WASTE
STORAGE AREA FOR
HIGH LEVEL WASTES
40m
10 m
10m
WASTE
CANISTERS
CRATON
SHIELDED WITH CERAMIC
(SYNROC ARTIFICIAL ROCK)

Fig. 3.

with numerical formulations. Different folding mechanisms operate in characteristic tectonic settings such as Flexural slip folding and Passive folding. We must avoid Passive folding mechanism because in this case deformation predominantly occurs in highly ductile rocks and especially in metamorphic terains which can extend to greater depths. But, in the case of Flexural slip folding, the time required for continuous rock deformation is quite long, at least of the order of $n \times 10^5$ years, and detachment seems to require a highly ductile layer at the base. This follows that the recumbent fold of which structural geology is evolved from Flexural slip folding mechanism can contain wastes as long as millions of years. Secondly, granatized rocks which are partly sedimentary and partly igneous should be considered for recumbent fold. Moreover, computer simulation experiments in view of Geotectonic and recent plate tectonic-theories should be conducted. We have to seek "superstructure" from the survey of geologically stable areas of Pakistan. A superstructure is formed due to shallow level of less mobile rocks (epeirogenic) uneffected by plutonic activity or metamorphism. Areas of crust which suffer no more than epeirogenic deformation are knwn as stable areas or cratons.

Hence, it follows from our discussions that recumbent folds and structurally stable areas known as cratons should be considered for the disposal of high level radioactive wastes.

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